

# PATENT COOPERATION TREATY

# PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

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Applicant's or agent's file reference PCT1880KG057	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/PEA/416)	
International application No. PCT/EP 03/06585	International filing date ( <i>day/month/year</i> ) 23.06.2003	Priority date ( <i>day/month/year</i> ) 28.06.2002
International Patent Classification (IPC) or both national classification and IPC B01J29/76		
Applicant HALDOR TOPSOE AS et al.		

- This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 6 sheets, including this cover sheet.  
  
☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 2 sheets.

- This report contains indications relating to the following items:
  - I ☒ Basis of the opinion
  - II ☐ Priority
  - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
  - IV ☐ Lack of unity of invention
  - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
  - VI ☐ Certain documents cited
  - VII ☐ Certain defects in the international application
  - VIII ☐ Certain observations on the international application

Date of submission of the demand  05.12.2003	Date of completion of this report  10.09.2004
Name and mailing address of the International preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer  Gosselin, D  Telephone No. +49 89 2399-8400  

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. **PCT/EP 03/06585**

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, Pages**

1-31 as originally filed

**Claims, Numbers**

1-10 received on 29.07.2004 with letter of 29.07.2004

**Drawings, Sheets**

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).  
☐ the language of publication of the international application (under Rule 48.3(b)).  
☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.  
☐ filed together with the international application in computer readable form.  
☐ furnished subsequently to this Authority in written form.  
☐ furnished subsequently to this Authority in computer readable form.  
☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

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5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes: Claims	
	No: Claims	1-10
Inventive step (IS)	Yes: Claims	
	No: Claims	1-10
Industrial applicability (IA)	Yes: Claims	1-10
	No: Claims	

2. Citations and explanations

**see separate sheet**

**Re Item V**

**Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Preliminary statement concerning the amended claims.
  - 1a. The catalyst used in the process according to claims 1 to 10 is defined by reference to acidity indexes, which are defined in the application. The definitions of the acidity indexes given in the application may be regarded as clear and concise. However, no meaningful comparison with the prior art can be made, since the catalysts known in the art for the same purpose are not commonly defined by reference to acidity indexes as defined in the application. Consequently, the subject-matter of claims 1 to 10 does not meet the requirements of Article 6 PCT.

Consequently, the examination has been restricted to a process according to claims 1 to 10 involving the use of the catalyst as defined in these claims with the acidity indexes having been omitted. If these features should be of importance it is the duty of the applicant to show differences between the compositions according to claims 1 to 10 of their application and at least all those compositions which are explicitly disclosed or can be unambiguously derived from the documents cited in the search report, in particular D8 and D6.

- 1b. The wording of claim 1 renders the claim as a whole unclear. In the application as originally filed the term "silica-alumina" was never used per se for defining the amorphous inorganic oxide material. "Silica and alumina and their combinations" were disclosed (claim 5 and page 13, lines 26-30 as originally disclosed). It is actually not clear what the applicant means by "silica-alumina and alumina and combinations thereof". A priori the combination of the two should be a silica-alumina comprising material so that the terms "combinations thereof" may be redundant. Eventually one should read "silica and alumina and combinations thereof" to be closer to the wording of the application as originally filed.
  - 1c. The restriction of the amorphous organic oxide material to "silica-alumina (or silica?) and alumina and combinations thereof" is de facto a selection among the list of oxides initially disclosed in claim 5 and page 13, lines 26-30 of the application. This specific selection among the list of original claim 5 was neither

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explicitly nor implicitly disclosed in the application as originally filed. Said application does not comprise evidence, which would lead the skilled man to make this selection. Accordingly the amended claims might be objectionable under Rule 70.2(c) PCT.

- 1d. Claims 9 and 10 does not meet the requirements of Article 6 PCT (conciseness). The claims are only referring to the standards for measuring the indexes of claims 2 and 3, and did not provide any limitation over the content of claims 2 and 3. The content of claims 9 and 10 should be respectively introduced into claims 2 and 3.
- 1e. The description must still be brought into conformity with the amended claims. References to the catalyst and/or to other reactions (i.a. pages 30-31) as subject-matter of the invention should be deleted.

2. Reference is made to the following documents:

D1: WO-A-03045548  
D2: WO-A-0117901  
D3: EP-A-1106592  
D4: US-A-5800698  
D5: WO-A-9613563  
D6: US-A-4612108  
D7: US-A-4812223  
D8: US-A-4486296  
D9: US-A-4601993  
D10: US-A-5011593

3. The present application does not meet the requirements of Article 33(1) PCT, because the subject-matter of claims 1 to 10 is not new in the sense of Article 33(2) EPC or does not involve an inventive step in the sense of Article 33(3) PCT in view of at least one of the following documents (cf: the passages quoted in the search report).
- 3a. The catalysts of the application (claims 1 to 10) would only differ from those of the cited prior due to specific acidity indexes. However it is the duty of the applicant to show that the claimed catalysts are really novel over all the compositions of the prior art cited in the search report. These citations are only a short selection among numerous documents; the introduction of further documents at a later

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stage of the examination is not excluded. The acidity indexes of catalysts of the cited documents was never determined according to the methods defined at pages 22 to 24 of the application. In view of initial objections, the subject-matter of the claims was restricted to the process for the preparation of middle distillate by selective conversion of hydrocarbon.

- 3b. The documents D1 and D3 to D10 disclose the use of the catalyst in hydrocarbon conversion processes in presence of hydrogen. It is admitted that D1 to D5, D7 and D9 to D10 are not concerned with a process according to claims 1 to 10 of the application.

However, D6 and D8, but more particularly D8, are concerned with the production of middle distillates (D6: Table 2; D8: column 10, lines 22-37). It is admitted that the beta zeolite of Example 1 of D8 is not precisely defined, however according to column 6, lines 25-39, it is clear the the silica to alumina ratio should be at least 100. Therefore, the embodiments A and B of Example 1 of D8 can only differs from the process of claim 1 in that the support have different acidity indexes NH<sub>3</sub>-Al and IEC-Al. Note that present claim 1 of the application does not exclude the presence of an additional X or Y zeolite in the catalytic composition.

The process according to claims 1 to 10 can be novel only if the novelty of the catalyst according to claims 1 to 10 is established.

4. Once the novelty will have been established, the applicant should establish the inventive step. The comparison with zeolite Y alone, or beta zeolite having a silica to alumina ratio of 25, is not relevant, since D8 (closest prior art) discloses a process for the preparation of middle distillates by selective conversion of hydrocarbon under hydrogenating conditions with a catalyst composition comprising a zeolite beta having a silica to alumina ratio of at least 100 with or without the presence of additional Y or X zeolite in the catalyst composition. The additional comparative examples filed by the applicant during the PCT examination did not meet this requirement (beta zeolite with a silica to alumina ratio of 25). Arguments based on the disclosure of US-A-5 447 623 cannot be considered, since this document is not the closest prior art, the beta zeolite referred to by the applicant having a silica to alumina ratio lower than 100.

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**NEW CLAIMS 1 TO 10**

1. Process for preparation of middle distillates by selective conversion of a hydrocarbon containing feedstock under hydrocracking conditions with a hydrocarbon conversion catalyst comprising one or more hydrogenation components supported on a support comprising a beta zeolite and an amorphous inorganic oxide, the beta zeolite having a  $\text{SiO}_2:\text{Al}_2\text{O}_3$  molar ratio of at least 50, and the amorphous inorganic oxide consisting of silica-alumina and alumina and combinations thereof, the support having an Ion Exchange Capacity-Acidity Index of less than 3.7.

2. Process of claim 1 wherein the support has an  $\text{NH}_3$ -TPD Acidity Index of less than 3.5.

3. Process of claim 1 or 2, in which the  $\text{NH}_3$ -TPD Acidity Index is less than 2.3 and/or the Ion Exchange Capacity-Acidity Index is less than 2.7.

4. Process of claim 1, wherein the beta zeolite has a  $\text{SiO}_2:\text{Al}_2\text{O}_3$  molar ratio of at least 100.

5. Process of claim 1, wherein the one or more hydrogenation components are selected from the elements of Group VIII and/or Group VI B.

6. Process of claim 5, wherein the hydrogenation components are selected from the group consisting of tungsten, molybdenum, nickel and combinations thereof.

7. Process of claim 6, wherein the hydrogenation components are a combination of nickel and tungsten.

8. Process of claim 1, wherein the support comprises less than 50 wt% zeolite beta and at least 50 wt% amorphous inorganic oxide.

9. Process of claim 1 or 3, wherein the Ion Exchange Capacity-Acidity Index of the ion exchange capacity of zeolite beta is normalized by the ion exchange capacity of a catalyst support containing standard zeolite beta having a  $\text{SiO}_2:\text{Al}_2\text{O}_3$  molar ratio of 25.

10. Process of claim 2 or 3, wherein the  $\text{NH}_3$ -TPD Acidity Index is the number of moles  $\text{NH}_3/\text{g}$  zeolite of zeolite beta normalized by the number of moles  $\text{NH}_3/\text{g}$  zeolite of a catalyst support containing standard zeolite beta having a  $\text{SiO}_2:\text{Al}_2\text{O}_3$  molar ratio of 25.